MASTER

PREPRINT UCRL- 78978

CONF-770575--4

Lawrence Livermore Laboratory

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7-1-77

THIS PAPER WAS PREPARED FOR SUBMISSION TO:
25TH ANNUAL CONFERENCE ON MASS SPECTROMETRY AND ALLIED TOPICS
AMERICAN SOCIETY FOR MASS SPECTROMETRY
MAY 29 - JUNE 3, 1977 WASHINGTON, D. C.

THE HALF-LIFE OF ^{239}Pu AS DETERMINED BY PULSE-COUNTING MASS SPECTROMETRY

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We have measured the half-life of ^{239}Pu by determining the ingrowth of ^{235}U during a known time by isotope dilution mass spectrometry techniques.

When ^{239}Pu decays, it gives off energetic α particles and forms ^{235}U. Writing it generally, $N \xrightarrow{\lambda} M + \alpha + E$, in which λ is the characteristic decay constant for the process. The relationship between λ and the desired half-life is $t_{1/2} = \ln 2/\lambda$. Solving the rate equation for short periods of time gives: $-\Delta N_{①}/N\Delta t = \Delta M_{②}/N\Delta t = \Delta \alpha_{③}/N\Delta t = \Delta E_{④}/N\Delta t = \lambda$. The classical radiochemist would use number ①; the calorimetrist, number ④; we mass spectrometrists chose number ②.

We received from LASL the master plutonium solution with a known number of ^{239}Pu atoms in it on which to do the half-life study. Some of the master solution was used to calibrate a high-purity ^{242}Pu tracer mass spectrometrically.

Five portions of the master solution, each containing approximately 10 mg of plutonium, were aliquoted for the growth experiment. ^{237}U tracer which had been calibrated by gamma-ray counting on a Ge(Li) diode was added to two of the five samples and a reagent blank. Each of the samples underwent purification by ion exchange to completely remove ^{235}U prior to the ^{235}U ingrowth period. After purification the samples traced with ^{237}U were found by gamma counting to contain less than 20 ppm of the ^{235}U present in the master solution.

Each of the five samples was placed in a volumetric flask, diluted to volume, and weighed. A small aliquot of this solution was weighed and diluted for assay with the ^{242}Pu tracer. This assay established the ^{239}Pu concentration of the purified solution. The remainder of the solution was transferred to a clean flask and weighed. ^{233}U tracer, prepared by Allied Chemical Corporation for the Safeguard Analytical Laboratory Evaluation Program (SALE), was added to each flask and the solution allowed to grow ^{235}U.

At the end of the growth period, the uranium was separated from plutonium by ion exchange and the time precisely recorded. The ratio of ^{235}U/^{233}U was measured mass spectrometrically on two mass spectrometers: MSXL a tandem 60°-sector mass spectrometer of 34 cm radius operating in the surface ionization mode; MSVIII a sequential dc single focusing 60° instrument of 34 cm radius also operating in the surface ionization mode.

Plutonium or uranium chloride was dried and calcined on the tantalum sample filament and mounted in the source of the spectrometer approximately 1 mm from the rhenium ionizing filament. In each system, the electron

multiplier output pulses were fed through a preamplifier into a multisumming scaler. A dedicated PDP-8 computer was programmed to abstract the information from the scaler, subtract the background, make multiplier coincidence corrections, and output the isotope ratios in a suitable format. Each sample filament was run in both spectrometers. In the case of the ^{242}Pu samples, mass bias corrections are self-cancelling. NBS uranium standards run in both spectrometers showed no mass bias for uranium.

We show here the results for one of the five samples as an example of the ^{239}Pu half-life determination. Table I shows the data for the assay of the dilution of MS-1 for ^{239}Pu using the ^{242}Pu tracer. Combining the average value of 3.71786×10^{14} ^{239}Pu atoms/gram with the dilution factor (56250.0) we obtain 2.09130×10^{19} atoms of ^{239}Pu in the MS-1 sample. Table II shows the quadruplicate measures of the uranium $\frac{235}{233}$ ratio in MS-1 at the end of the growth period. Combining this ratio with the known amount of tracer added (2.80858×10^{14} ^{233}U atoms) we obtain the value of 3.68430×10^{14} atoms of ^{235}U formed.

$$t_{1/2} = \frac{(0.693147)(223.8175 \text{ days})(2.09130 \times 10^{19} \text{atoms})}{(3.68430 \times 10^{14} \text{ atoms})(365.243 \text{ days/year})} = 24{,}110.1 \text{ yrs.}$$

Table I

QUADRUPLICATE PLUTONIUM ASSAY SAMPLES FROM MS-1

Sample ID	^{239}Pu atoms/g of MS-1 dilution
MS-1-1	$3.71580 \times 10^{14} \pm 0.00026 \times 10^{14}$ [a]
MS-1-2	$3.71734 \times 10^{14} \pm 0.00020 \times 10^{14}$
MS-1-3	$3.71518 \times 10^{14} \pm 0.00039 \times 10^{14}$
MS-1-4	$3.72311 \times 10^{14} \pm 0.00039 \times 10^{14}$
AVERAGE	$3.71786 \times 10^{14} \pm 0.00181 \times 10^{14}$

Table II

RATIO OF $\frac{235}{233}$ IN MS-1

Run #	Ratio $\frac{235}{233}$ Uranium
XL-1-1	1.31123 ± 0.00026 [a]
XL-1-2	1.31072 ± 0.00020
XL-1-3	1.31292 ± 0.00040
MSVIII-1	1.31216 ± 0.00059
AVERAGE	1.31176 ± 0.00049

[a] Errors on ratio are one standard deviation based on counting statistics.

Table III

Sample Identification	Half-life in Years
MS-1	24,110.1
MS-2	24,128.2
MS-3	24,070.6
MS-4	24,071.9
Q -2	24,065.9
AVERAGE	$24{,}089.3 \pm 12.5$

Estimating systematic errors and adding them in quadrature to the random errors calculated gives for the half-life of ^{239}Pu 24,089 ± 22 years.

Work performed under the auspices of the U.S. Energy Research & Development Administration under contract No. W-7405-Eng-48.